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Potential to reduce the concentrations of short-lived climate pollutants in traffic environments: A case study in a medium-sized city in Brazil

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ABSTRACT

Understanding the spatio-temporal variation of short lived climate pollutants (SLCP) over cities is critical to protect public health and mitigate climate change. There is a lack of knowledge about the distribution of SLCP in South America, mainly in medium-sized cities (< 1 million inhabitants). This work reports on results of a campaign conducted at several sites (street canyon, urban background, and suburban) in Londrina, Brazil. The pollution datasets (black carbon BC, nitrogen oxides NOx, ozone O_3, fine particles $PM_{2.5}$, and particle number PN) were analyzed by using descriptive statistics, diurnal cycles, time-space correlations, and establishing linkages with traffic rates and with wind conditions. Hourly mean (± standard deviation) BC concentrations ranged from 0.58 (± 0.54) in the suburban area to 3.43 (± 2.69) $\mu g\,m^{-3}$ at the street canyon site (10,150 vehicles d^{-1} on weekdays), showing a high spatio-temporal variability even at short scales. PN (mean of 17,469 cm^{-3}) and NOx (mean of 51.58 $\mu g\,m^{-3}$) concentrations tracked BC levels and traffic rates, particularly the number of diesel buses at the canyon site. $PM_{2.5}$ and O_3 levels presented a lower spatial variability, with poor correlation with traffic rates in the canyon and were more tied to regional sources. NOx, BC, and PN showed the highest reduction potentials connected to the abatement of traffic emissions in the city center, particularly by targeting heavy-duty diesel vehicles. The reduction of $PM_{2.5}$ and O_3 concentrations is more challenging due to a significant regional contribution that requires cutting down emissions at state, national or even transboundary scales.

1. Introduction

The Paris Agreement entered into force in November 2016 with the main goal of limiting global warming to below 2 °C compared to pre-industrial levels (UNFCCC, 2015). To reach this ambitious target, 178 countries (as of June 2018) submitted national climate action plans to decrease greenhouse gases (GHG) emissions as part of a global coordinated action. However, this strategy has to be complemented with the reduction of emissions of the so-called short-lived climate forcers (SLCF) to slow down global warming in the near time (UNEP, 2017).

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SLCF are compounds that modify the atmospheric energy budget typically within the first decade after their emission, and include methane (CH_4), some hydrofluorocarbons (HFC), tropospheric ozone (O_3), and aerosols such as black carbon (BC), organic carbon (OC) and $PM_{2.5}$. The precursors of O_3 and $PM_{2.5}$ are also considered, including carbon monoxide CO, nitrogen oxides NOx, non-methane volatile organic compounds NMVOC, sulfur dioxide SO_2, and NH_3 (IPCC, 2018). Curbing the emission of SLCF has also co-benefits for air quality, human health and ecosystems because some SLCF are harmful air pollutants (WHO, 2015), and dubbed short-lived climate pollutants (SLCP).

In that context, some international organizations, such as the Climate and Clean Air Coalition (CCAC) and the International Council on Clean Transportation (ICCT), have been promoting the reduction of SLCP in various economic sectors (transport, industries, agriculture, waste management, and residential cooking and heating) by helping create policies and practices that target these activities. In May 2018, even the International Panel on Climate Change (IPCC) –which has traditionally focused on the reduction of GHG– organized an expert meeting to explore the need for methodologies to develop SLCF inventories, to assess the existing observations of SLCF around the world, and to estimate the impacts of their emissions on the climate. These international actions are also reflected at national level in certain developed countries, such as the states of the European Union, and very recently (as of June 1st 2018), the U.S. Climate Alliance committed to reducing SLCF in the entire country.

Brazil is a continent-sized country, ranking 5th in surface area and human population (210 million inhabitants), and has the 4th largest on-road vehicle fleet in the world (98 million, DENATRAN, 2018). In terms of GHG emissions, Brazil is the largest GHG emitter in Latin America and, as of 2016, the 6th largest in the world (Janssens-Maenhout et al., 2017). Moreover, according to the Integrated Assessment of SLCP in Latin American and the Caribbean (UNEP, 2016), Brazil was the largest SLCP emitter in the region in 2010. Hence, as a complementary measure to meet the Paris Agreement and encouraged by the significant health and climate benefits of reducing SLCP emissions, Brazilian environmental agencies should measure SLCP concentrations and their spatial distribution and identify their source contributions. However, monitoring SLCP concentrations in Brazil is still challenging due to financial and infrastructure constraints, limited number of stations, lack of instrument maintenance, spare parts and technical expertise (Instituto de Energia e Meio Ambiente, 2014). Thus, there is an urgent need to fill a knowledge gap in terms of local SLCP emissions sources and their impact on the ambient concentrations in cities, where most of the population lives (85%).

Different from Europe and North America, where most studies of traffic-related pollutants have been carried out (e.g., Buzorius et al., 1999; Birmili et al., 2015; Blanchard et al., 2014; Kutzner et al. 2018; Hagemann et al., 2014; Krecl et al., 2014; Dos Santos-Juusela et al., 2013; Johansson et al., 2007), Brazil has no regular vehicle inspection and maintenance (I/M) programs in place at the moment. This triggers the presence of the so-called gross emitters that are usually old heavy-duty diesel vehicles, with no maintenance at all, and that can largely impact the ambient concentrations of SLCP particularly on busy streets. Another unique aspect in Brazil is the extended use of biodiesels; gasoline is blended with 27% of anhydrous ethanol by volume (E25), and the automotive diesel contains 7% of biodiesel with sulfur content of 500 ppm or 10 ppm (the latter for vehicles manufactured from 2012). In terms of vehicle technologies, the Brazilian fleet is rather old (mean of 9.3 years with 49% of vehicles between six and 15 years old; Sindipeças, 2017), emission standards lag about four to five years behind EURO standards, and the use of diesel particulate filters (DPF) was never adopted. In the case of EURO-V equivalent buses and trucks, NOx emissions in Brazil can be higher than certification levels because of inadequate use of the urea-based after-treatment technology to reduce NOx emissions and the low exhaust temperatures to activate the selective catalytic reduction system when circulating in urban areas with frequent stop-and-go driving (Façanha, 2016).

Studying the spatio-temporal distribution of air pollutants helps (i) characterize their emission sources, which is key information for successful strategies to protect public health and to mitigate climate effects, (ii) diagnose the population exposure to harmful species, (iii) understand their lifecycle in the urban atmosphere, and (iv) validate air quality dispersion models. The spatio-temporal variability of traffic-related pollutants has been traditionally studied by gathering data from monitoring sites across cities over several months or even years (e.g., Birmili et al., 2015; Blanchard et al., 2014; Kutzner et al. 2018), or during short-term campaigns for research purposes (e.g., Buzorius et al., 1999; Gu et al., 2013; Dos Santos-Juusela et al., 2013; Krecl et al., 2018, 2014, 2011). The introduction of portable instruments in the market over the last decade favored the flourishment of mobile sampling studies with great detail in urban areas, particularly highlighting the large variability of the ambient concentrations of BC and $PM_{2.5}$ (e.g., Krecl et al., 2014; Targino et al., 2016), particle number PN and NOx (e.g., Hagemann et al., 2014) in connection to vehicular sources.

Besides road traffic and other emission sources, meteorological conditions impact the spatio-temporal distribution of aerosol particles and gaseous compounds. For example, ventilation induced by high wind speeds (WS) prevents the local build-up near the sources, but contributes to long-range transport of pollutants especially during persistent wind conditions. Winds can also have a strong interaction with urban configurations, particularly within street canyons. Rooftop winds blowing perpendicular to the street and $WS > 1.5\,m\,s^{-1}$ usually trigger a recirculation vortex, causing higher concentrations of air pollutants in the leeward side (Hunter et al., 1992; Krecl et al., 2015).

To implement successful strategies and actions to reduce air pollution levels in cities, it is of outmost importance to quantify local and regional source contributions. Several approaches were developed over the years to calculate these contributions for all sectors or even for specific activities like traffic emissions. To this end, some studies used complex dispersion models at different spatial scales (e.g., Gidhagen et al., 2018; Pecorari et al., 2012; Sciare et al., 2010) and others estimated the contributions based on measurements conducted at key locations in the city and on its outskirts (e.g., Ketzel et al., 2004; Imhof et al., 2005; Krecl et al, 2011; Clapp and Jenkin, 2001).

This study analyzes the spatio-temporal variability of the concentrations of several SLCP (BC, O_3, NOx, $PM_{2.5}$) plus PN in a medium-sized Brazilian city (554 thousand inhabitants) by using data measured within a trafficked street canyon, at a rooftop site and on the city's outskirts. We highlight the importance of including PN in this research, since traffic environments are largely

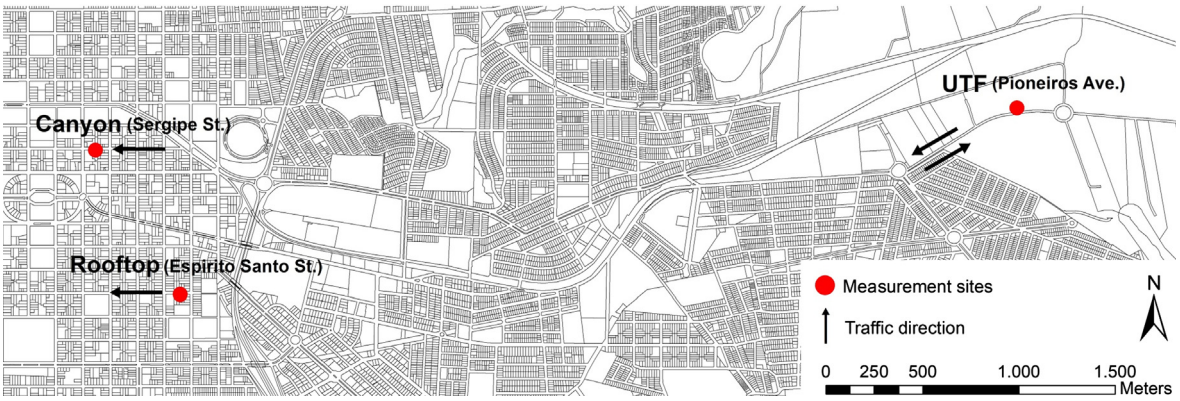

Fig. 1. Map showing the location of the street canyon, rooftop and university campus (UTF) sites.

dominated by ultrafine particles (diameter smaller than 100 nm) which can contribute little to $PM_{2.5}$ mass concentrations but pose serious health risks (HEI, 2013). We present descriptive statistics, diurnal patterns, spatial and temporal correlation analyses, and relationships with WS and wind direction (WD), since these are key variables influencing pollution dispersion. Finally, we calculate and discuss the urban and regional contributions to the atmospheric pollutants concentrations and the potential for their reduction.

2. Methodology

2.1. Measurement sites

The sampling campaign was conducted in the city of Londrina, located in the northern part of the state of Paraná (latitude: 23.29° S, longitude: 51.17° W). We set up four measurement sites: on both sides of a one-way street canyon, at a building rooftop (representative of urban background) and at a suburban site on the city's outskirts (Fig. 1). The traffic within the street canyon was organized into two lanes and managed by traffic lights situated at both ends of the transect (for a street canyon map and picture, refer to Krecl et al., 2016). Even though the vehicle speed limit (VS) was 40 km h^{-1}, start-stop driving was the general driving pattern and frequent congestions were observed within the canyon at rush hours.

The rooftop site was installed in the city center, on a one-way street (Espírito Santo St.) distant 800 m southeast from the street canyon location. The street has low traffic rate (TR, 140 veh h^{-1} in the morning and afternoon traffic peaks), and is 80 m from a more trafficked street (Brasil St., 800 veh h^{-1} at rush hours). The suburban site was located at the university campus (UTF), about 4.2 km from the canyon transect on the eastern edge of the city, with the closest neighborhood consisting of sparsely built detached houses about 400 m away, and ~30 m from the main road (3,100 veh d^{-1} and 300 veh h^{-1} in the evening peak).

2.2. Instrumental set-up

The instruments used in the sampling campaign were described in Krecl et al. (2018) and only a brief overview will be given here.

Table 1
Technical details of the air pollution monitors installed at the four sites.

Site	Pollutant	Model (manufacturer)	Flow rate [L min^{-1}]	Frequency [min]	Inlet height [m]	Period [year 2016]
Canyon South	BC	Microaeth AE51 (AethLabs, USA)	0.05	1	7.0	January 26th-March 27th
	O_3	Monitor 106M (2B, USA)	1.0	1	7.0	January 26th-March 27th
	$PM_{2.5}$	DustTrak (TSI, USA)	1.7	1	7.0	January 26th-March 27th
	NOx, NO	Monitor 42i (Thermo Sci., USA)	0.7	1	7.0	February 13th-March 27th
	N	CPC 3772; diluter 3332 (TSI, USA)	1.0	1	7.0	February 18th-March 27th
Canyon North	BC	Microaeth AE51 (AethLabs, USA)	0.05	1	4.8	January 26th-March 7th
Rooftop	$PM_{2.5}$	DustTrak (TSI, USA)	1.7	1	21.0	January 26th-March 7th
	BC	Aethalometer AE42 (Magee Sci., USA)	5.0	2	21.0	January 26th-March 7th
UTF	O_3	Monitor 49i (Thermo, USA)	1.0	1	1.2	January 26th-March 27th
	$PM_{2.5}$	DustTrak (TSI, USA)	1.7	1	3.0	January 26th-March 7th
	BC	Microaeth AE51 (AethLabs, USA)	0.05	5	3.0	January 26th-March 7th

Table 1 shows the technical details of the instrumental set-up. BC concentrations were measured with a benchtop aethalometer model AE42 operating at seven wavelengths (370, 470, 525, 590, 660, 880, and 950 nm) and with portable aethalometers model AE51 at 880 nm. A collocation experiment with all aethalometers in ambient air during three days showed a very high Pearson correlation coefficient R (R > 0.91 between the AE42 and the AE51 units, and R = 0.99 among the AE51 monitors) with regression slopes ranging between 0.92 and 0.99 for hourly data. A compensation for the loading effect was applied to the BC data following Virkkula et al. (2007) and Targino and Krecl (2016). The data was not corrected for scattering artifacts due to the lack of concurrent measurements of the aerosol scattering coefficient.

O_3 measurements came from two instruments that use the photometry principle, in which the concentrations were deduced from the absorption of radiation at 254 nm by O_3 molecules. Instrumental collocation yielded excellent agreement between the instruments for hourly measurements (R = 0.94, slope of 1). We report O_3 concentrations as mass concentration considering atmospheric pressure of 1013.25 hPa and air temperature of 25 °C. $PM_{2.5}$ concentrations were determined with DustTrak photometers operated with a 2.5-μm impactor fitted to the inlet, and subsequently corrected with a factor of 0.52 based on previous concurrent gravimetric analysis conducted at the UTF site (not published). The DustTrak monitors were collocated at the canyon site and the data were highly correlated (R = 0.99, slopes between 0.93 and 0.97).

PN concentrations were measured with a condensation particle counter (CPC 3772) coupled with a 1:100 dilution system (model 3332–100). The CPC 3772 has a 50% lower cut-off size of 10 nm and maximum measurable concentration of 10^6 cm^{-3}. To assess possible particle losses induced by the dilution system (Kulkarni et al., 2011), we collocated the CPC 3772-diluter suite with a CPC 3007 (TSI, USA) at the canyon South site. The instruments showed excellent correlation (R = 0.99) for 1-min concentrations in the range from 3,500 to 85,000 cm^{-3}, but the CPC 3007 showed higher values (up to 17%), which has been attributed to the different working fluids and different counting efficiency curves for particle diameter D_p < 20 nm (Krecl et al., 2018). However, the differences in PN concentrations between the CPC 3772 set-up and the CPC 3007 were within concentration accuracies reported by the manufacturer (± 20% and ± 10%, respectively). A NOx analyzer, based on the chemiluminescence principle, measured NO and NO_x at the canyon South site. We report NOx as NO_2 equivalent, at atmospheric pressure of 1013.25 hPa and air temperature of 20 °C.

We continuously recorded traffic data at the canyon site with a video camera (IP VIP S3020, Intelbras, Brazil), and thereafter hourly TR were manually extracted for the heavy-duty (HDV, trucks and buses) and light-duty (LDV, passenger cars, light-commercial, and motorcycles) vehicles. Meteorological variables were observed at the university campus with a temporal resolution of 1 min with an air temperature and relative humidity sensor (Vaisala model HMP45C, Finland), WS and WD sensors installed at a height of 10 m (Young model 05103, USA), pyranometer (Kipp & Zonen model CMP3, The Netherlands) and tipping bucket rain gage (Campbell Scientific model TB4, UK). Because the meteorological site was located on a relatively flat terrain outside the urban area and uninfluenced by the presence of large buildings, we assumed that the wind data provided a reasonable estimation of the wind above the pollutant monitoring sites depicted in Fig. 1.

2.3. Data treatment

All data were inspected for anomalous values, and negative concentrations (measurements lower than 2-standard deviation (SD) of the instrumental noise value) and obvious outliers were removed. Then, hourly averages were calculated for pollutant concentrations and meteorological variables from at least 45 min of valid measurements. All times are reported in local winter time (UTC-3) and as 24-h notation.

Temporal correlations were calculated with the Pearson correlation coefficient for one compound measured at different stations (intersite) and for different compounds monitored at the same station (intrasite). The spatial variation per compound was assessed by the coefficient of divergence (COD) following Wongphatarakul et al. (1998), in which values approaching zero indicate poor spatial variability, whereas values close to one are interpreted as high spatial variability.

For each site and pollutant, mean hourly concentration values were sorted by WS into 1-m s^{-1} intervals (except for the last bin that spanned from 5 to 8 m s^{-1} to enlarge the number of observations) and the mean concentration was calculated for each band. Concentration roses were calculated by averaging hourly pollutant concentrations for each WD sector (22.5° intervals). To facilitate comparison between pollutants measured at the same site, the concentration roses were normalized to the maximum value within each WD sector.

2.4. Urban and regional contrib utions

To estimate the contribution of the urban sources relative to the background levels, we calculated mean concentrations at the different sites relative to the concentrations at the urban background site following Ketzel et al. (2004) and Krecl et al. (2011). This methodology assumes that (i) the rooftop site is representative of the urban background conditions, and (ii) the suburban site is not influenced by the city emissions. The rooftop site chosen for this study is considered representative of the urban background pollution level in Londrina, based on the comparison of the mean BC concentrations at this site and at another rooftop site sampled in a previous campaign in 2014 (0.93 μg m^{-3} in 2014 vs. 0.88 μg m^{-3} in 2016; more details in Krecl et al., 2016 and Section 3.1). The second assumption is fulfilled for all pollutants except for O_3, due to the non-negligible correlation between O_3 concentrations at the street canyon and UTF sites (see Section 3.3).

Because our target is traffic environments, the street canyon is a suitable location for air quality monitoring since this configuration is located mostly in the inner city, where population and traffic densities are high. This combined feature enhances the exposure of residents to harmful atmospheric pollutants and poses higher mortality and morbidity risks (Forsberg et al., 2005). Given

that the traffic intensity is usually higher on weekdays (and this is the case for our study, see Section 3.2), we used weekday concentrations to estimate the impact of traffic contributions to the urban air quality.

In the case of O_3, we applied the methodology developed by Clapp and Jenkin (2003) that estimates the regional contribution of O_3 by calculating the independent term of the linear regression equation between the daylight mean mixing ratios of the oxidant ($OX = O_3 + NO_2$) and NOx. The daylight period was considered between 08:00 and 18:00 and for the entire sampling period at the canyon South site when NOx, NO_2 and O_3 were simultaneously available.

2.4.1. Determination of urban background concentrations

Because PN and NOx were not measured at the rooftop site, we estimated their mean background concentrations by two simplified approaches: (i) assuming a constant urban background value along the day calculated as the mean concentration of NOx (or PN) on weekdays during the early hours, when traffic was minimal, (ii) assuming that the same mean BC ratio between South canyon and rooftop sites holds for PN and NOx concentrations. Then, we compared our results with typical road-to-roof ratios of NOx and PN retrieved from the literature.

3. Results and discussion

3.1. Overall description

Daily mean air temperature and relative humidity varied between 20.6 °C and 29.8 °C, and between 61% and 96%, respectively. The mean WS was 2.7 m s^{-1} and the prevailing WD were south (19%), southeast (17%) and east (16%) (Fig. 5d). The accumulated rainfall over the study period was 314.2 mm, and it rained on 24 out of 62 days, with the largest daily accumulated amount of 63.2 mm on February 23rd. The mean daily solar energy dose was 18.5 MJ m^{-2}, varying between 5.7 (on February 23rd) and 27.4 MJ m^{-2} (on February 6th); the mean maximum solar irradiance was 842 W m^{-2} with a minimum of 323 (on February 23rd) and a maximum of 1113 W m^{-2} (on February 2nd).

To illustrate the spatio-temporal variability of the pollutant concentrations, Fig. 2 displays hourly values for a five-day period between February 19th and 23rd (Friday to Tuesday) when data coverage was maximum for all variables. The concentrations of PN, NOx and BC peaked inside the canyon (Fig. 3c, g, e) in the morning and evening on weekdays, whereas concentrations were much lower and had a flatter pattern on Sunday. BC concentrations were highest at the canyon North site reaching 19.5 μg m^{-3} on February 22nd at 18:00, while the other sites (canyon South, rooftop and UTF) reported 9.0, 2.1 and 0.9 μg m^{-3}, respectively (Fig. 3e, f, h, j). $PM_{2.5}$ concentrations showed a less pronounced intersite variation than BC, and were larger at the central sites except for a peak (21.2 μg m^{-3}) observed at UTF on Sunday at 10:00. Ozone patterns were very similar at the canyon South and UTF sites (Fig. 3i-j) with maximum concentrations in the early afternoon. Even though February 23rd was the rainiest day, the concentrations of all pollutants were higher on this day than on Sunday, indicating that the rain did not cause a significant washout of pollutants. Krecl et al. (2011) also observed that rainfall did not affect BC concentrations substantially at several sites in Stockholm.

Descriptive statistics of all pollutants were calculated for the period between January 26th and March 7th 2016 (Table 2). Missing data corresponds to time periods when the instruments were not operating due to maintenance, data download, flow rate or signal out of range.

Mean BC concentrations (at 880 nm) ranged from 0.58 to 3.43 μg m^{-3}, and SD between 0.54 and 2.69 μg m^{-3}. As expected, the highest BC concentrations were found at the canyon sites whereas the lowest levels were recorded at UTF, located on the city's outskirts. As pointed out by Krecl et al. (2016), the higher BC concentrations at the canyon's North façade compared to the South were due to the vortex effect and the proximity to a bus stop (see Section 3.4 for a detailed discussion). The largest intersite difference was observed for the 95th percentile, being six-fold higher at the North façade than at the UTF site, and the smallest differences were found for the 5th percentile (0.14–0.36 μg m^{-3}).

BC concentrations at the rooftop were slightly higher at 370 nm (mean of 0.97 μg m^{-3}, not shown) than at 880 nm wavelength (0.88 μgm $^{-3}$), suggesting the presence of organic carbonaceous particles, known for having stronger absorption towards the UV band (Moosmüller et al., 2009). The median absolute deviation (MAD) –a robust measure of statistical dispersion and less sensitive to outliers– was calculated as the median of the absolute difference between each individual observation and the dataset median. Values of MAD for BC ranged from 0.32 to 2.05 μg m^{-3}.

$PM_{2.5}$ concentrations were also higher within the street canyon than at the other two sites, but presented smaller intersite variability than BC levels (for example, the 95th percentile at the canyon station was only 1.5 times higher than at the UTF site). The BC to $PM_{2.5}$ ratios were 0.33 at the canyon site, 0.17 at the rooftop, and 0.11 at the suburban station, indicating a decreasing contribution from the motorized traffic to the $PM_{2.5}$ fraction. On average, O_3 concentrations were slightly higher at the canyon site when compared to the suburban station, and the diurnal patterns per day of the week should be analyzed for a better insight (Section 3.2, items x and xi).

In the case of PN and NOx, the instruments were installed later, reducing the number of samples for the coincident period. Because of this lag, we decided to extend the measurement period at the canyon South site until March 27th. Nevertheless, the descriptive statistics of PN and NOx concentrations were very similar when comparing the periods February 13rd to March 7th and February 13rd to March 27th. For example, mean PN concentrations were 17,469 vs. 17,161 cm^{-3}, and NOx were 51.58 vs. 52.81 μg m^{-3}, respectively. PN and NOx presented a large temporal variability, which is consistent with data collected at curbside sites, with peak values matching high daytime TR and cleaner nighttime periods.

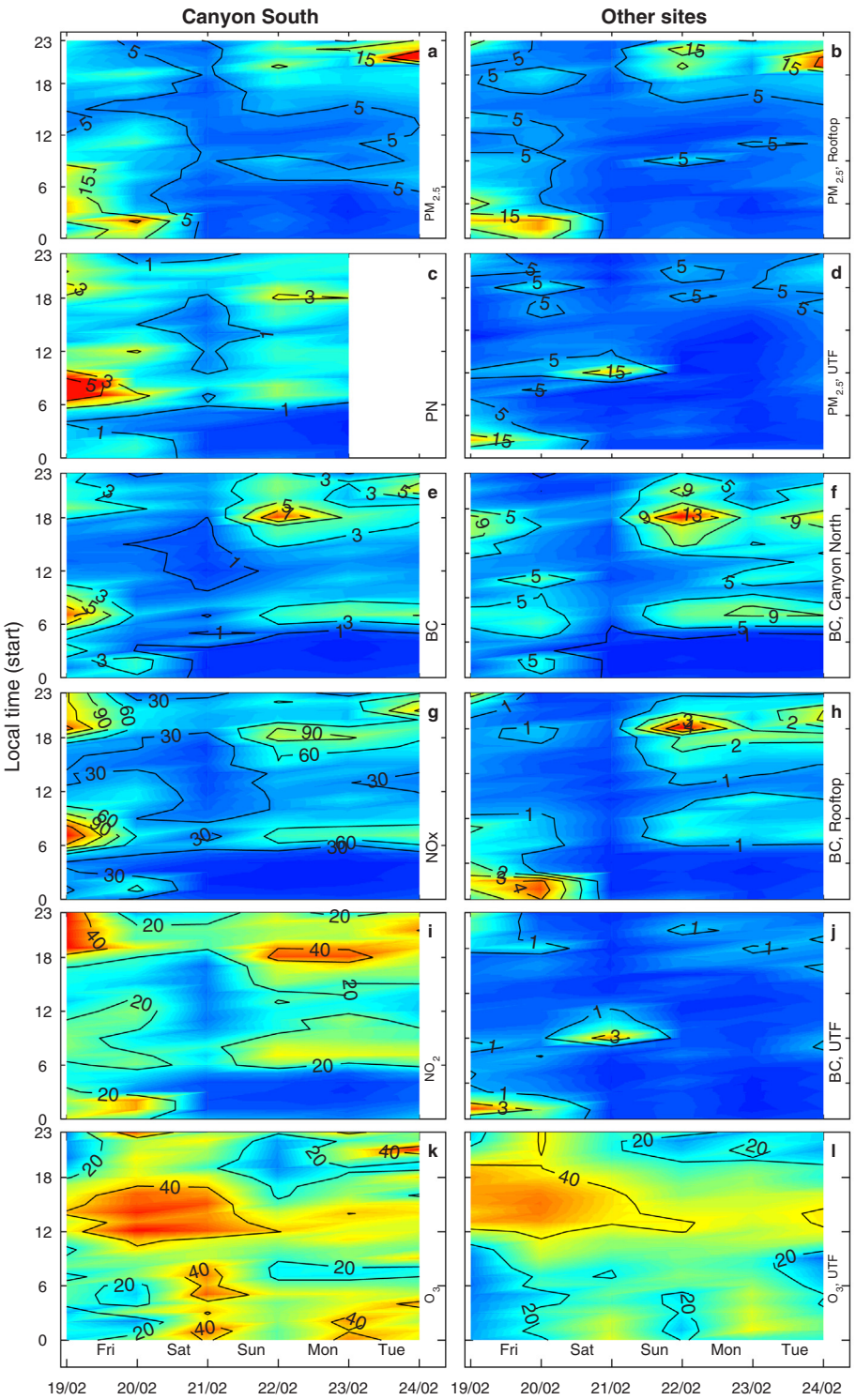

Fig. 2. Five-day hourly concentrations of pollutants for the canyon South site (left panels) and other sites (right panels) expressed in [μg m^{-3}], except for PN [10^4 cm^{-3}]. Note that subpanels e, f, and h present different color scales for BC concentrations.

3.2. Traffic and air pollution patterns

On average, the canyon street carried 10,150, 8,700 and 3,500 veh d^{-1} on weekdays (Monday through Friday), Saturday and Sunday (including public holidays), respectively. Matching the time of intense commercial activity in the city center, TR peaked in

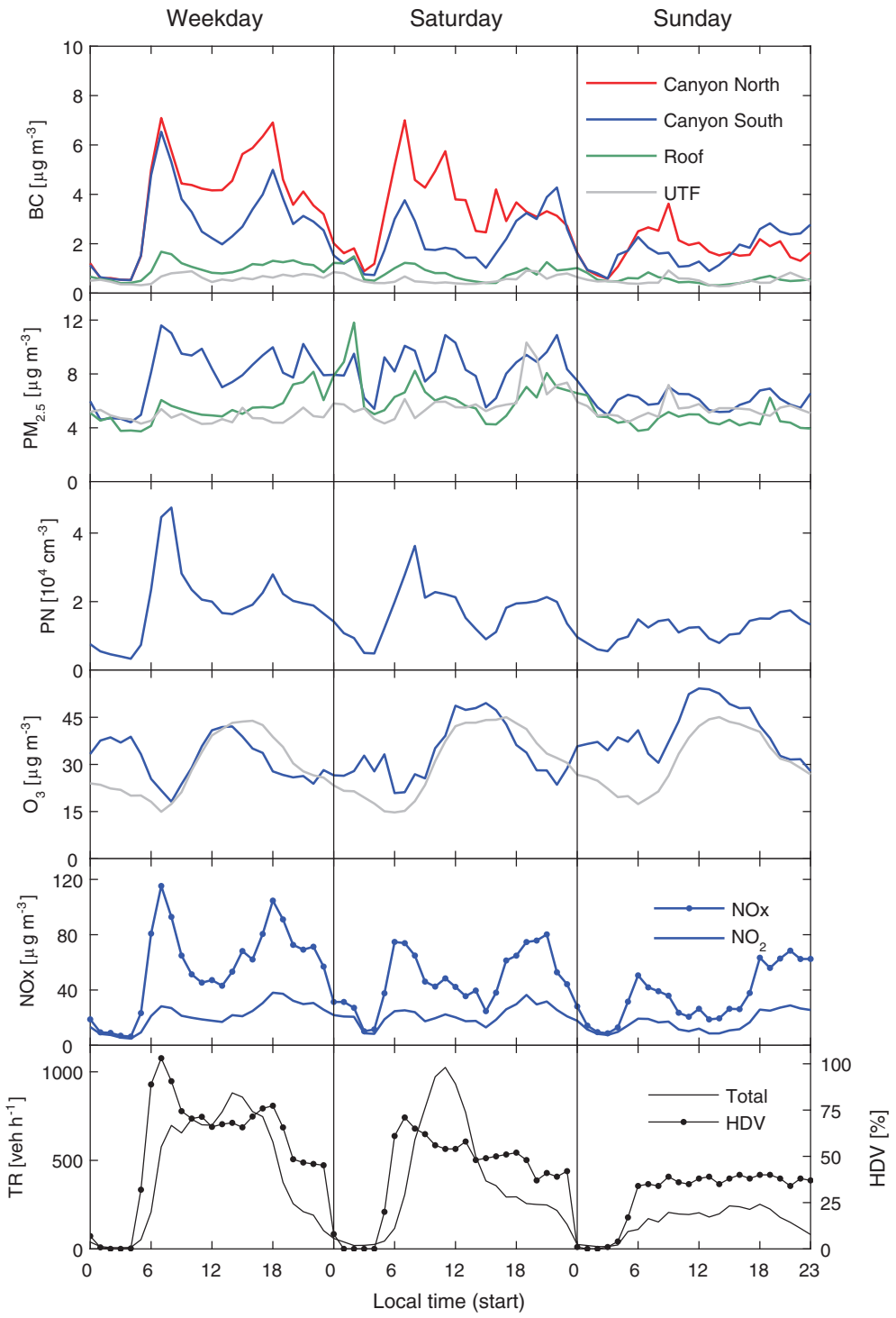

Fig. 3. Mean diurnal cycles of several pollutants and traffic rates (total and HDV) for weekdays, Saturdays and Sundays in the period January 26th - March 7th 2016.

the early afternoon on weekdays, and at 11:00 on Saturdays (Fig. 3). Sundays showed a reduced TR variation and the maximum values occurred in the late afternoon. The HDV fleet rate was highest in the early morning at weekdays and was dominated by public buses, all powered by diesel fuel (more details in Krecl et al., 2018).

Because of the different traffic patterns observed along the week, the diurnal cycles of the pollutant concentrations were calculated separately for weekdays, Saturdays and Sundays (Fig. 3). At the canyon sites, the concentrations were higher on weekdays

Table 2
Descriptive statistics of hourly pollutant concentrations in the period January 26th to March 7th, 2016.

Pollutant and site	Units	Mean	SD	Median	MAD	Percentile				Samples (#)
						5°	25°	75°	95°	
BC_{North}	[μg/m^3]	3.43	2.69	2.91	2.05	0.36	1.35	4.82	8.58	999
BC_{South}		2.55	1.99	1.97	1.51	0.35	1.13	3.41	6.49	1004
[a]BC_{Roof}		0.88	0.69	0.71	0.47	0.20	0.43	1.08	2.12	982
BC_{UTF}		0.58	0.54	0.44	0.32	0.14	0.27	0.71	1.42	979
$PM_{2.5South}$	[μg/m^3]	7.70	4.15	7.04	3.09	2.57	4.84	9.62	15.77	1006
$PM_{2.5Roof}$		5.25	3.44	4.71	2.32	1.24	3.19	6.57	10.81	875
$PM_{2.5UTF}$		5.06	3.23	4.56	2.22	1.27	3.06	6.45	10.27	941
[b]O_{3South}	[μg/m^3]	33.46	13.65	32.05	10.83	13.08	24.20	41.23	57.88	863
[b]O_{3UTF}		29.85	13.49	27.86	11.82	10.58	19.82	39.57	53.12	1004
PN_{South}	[cm^{-3}]	17,469	12,781	15,837	9,134	2,631	8,665	22,503	40,470	517
[c]NOx_{South}	[μg/m^3]	51.58	39.44	41.67	30.89	4.65	22.61	72.84	129.29	441
NO_{2South}	[μg/m^3]	20.72	11.66	20.20	9.28	4.00	12.01	27.46	42.56	441

[a] 880 nm.
[b] Expressed at T = 25 °C, P = 1013.25 hPa. [c]Expressed as NO_2 at T = 20 °C, P = 1013.25 hPa.

compared to Saturdays and Sundays for all species but $PM_{2.5}$ and O_3. The rooftop and UTF sites showed flatter patterns during the day and along the week for particulates, indicating a reduced impact from traffic emissions when compared to the canyon sites. Because of diurnal changes on the dilution rate due to meteorology and traffic-generated turbulence (Krecl et al., 2018), ventilation was favored during daytime (08:00–18:00) rather than at nighttime. The key features of the diurnal cycles are:

i. On weekdays, BC revealed two peaks at the canyon sites (07:00 and 18:00) matching the times of the day with highest number of HDV. Similar to results by Krecl et al. (2016) within the same canyon in 2014, no difference in BC concentrations between the North and South façades was found until 09:00. However, a significant difference was observed in the period 10:00–18:00 in connection with frequent buses stopping at a bus stop closer to the North site (20 m). The queues for dropping off and picking up passengers caused reduced overall VS.
ii. On Saturdays, BC concentrations at the North side were statistically higher than at the South side in the period 07:00–16:00 (Mann-Whitney *U* test at 95% confidence level). Besides being further from the bus stop (52 m), a 2-m width parking lane prevents vehicles from circulating close to the South façade when that lane is occupied (until the end of the commercial activity).
iii. On Sundays, BC levels did not show significant differences between the North and South sides. This behavior might be explained by a combination of factors: (a) lower total and HDV TR when compared to other days, (b) lower street occupancy that led to higher VS, (c) the parking lane was rarely occupied and vehicles could also drive closer to the South façade, and (d) fewer buses stopped because of lower number of passengers and reduced service.
iv. The $PM_{2.5}$ concentrations at the South façade followed the same pattern as BC, but for a second maxima on Saturdays matching the period of highest total TR (10:00–11:00). This second peak could have large contributions from dust resuspension that impacted the $PM_{2.5}$, but not the BC fraction.
v. $PM_{2.5}$ cycles at the background sites showed no clear resemblance with the cycle within the canyon site on weekdays and Saturdays.
vi. Between 00:00 and 03:00, the canyon and rooftop concentrations of BC, $PM_{2.5}$, PN and NOx were higher on Saturdays than on weekdays and Sundays and were attributed to traffic in connection to nightlife around the monitoring sites, particularly close to the rooftop site.
vii. At the canyon South, PN, NOx and BC concentrations tracked one another closely, showing the strong relationship among these traffic-related pollutants and the consistency in the dataset. Periods with high concentrations matched the hours with high percentage of HDV traffic and lower dilution rate values (not shown).
viii. The BC diurnal cycle at UTF was fairly invariant on Saturdays and Sundays, and a modest peak was observed between 06:00 and 12:00 in response to the vehicular traffic. The rooftop site showed faint peaks of BC concentrations on weekdays and Saturdays matching the times of peaks at the canyon site.
ix. At UTF, no significant differences were observed for the O_3 diurnal cycles when comparing weekdays, Saturdays and Sundays. O_3 peaked in the early afternoon, in response to the photochemical processes and started to decline when loss mechanisms –dry deposition on the ground and scavenging by NO titration (Monks et al., 2015)– dominated, reaching a minimum at sunrise.
x. O_3 measurements conducted at the canyon site showed a secondary maximum in the early hours. Since production ceases at night, studies reporting a nighttime O_3 peak attributed this pattern to horizontal advection or vertical transport of ozone-enriched air (e.g., Kulkarni et al., 2013; Mavrakis et al., 2010). Because this increase was not observed at the UTF site, we hypothesize that the transport might have been linked to local scale circulation. On top of the transport mechanism, O_3 was partially removed by NO titration on Saturdays between 00:00 and 03:00 (see vi), when NO concentrations were not negligible.
xi. The canyon site also exhibited the so-called “ozone weekend effect” (e.g., Fujita et al., 2003) with higher O_3 concentrations at weekends than on weekdays, particularly on Sundays. Lower NO concentrations and higher NO_2/NOx ratios on Saturday and

Table 3
R and COD between the same pollutant but measured at different sites.

	R [-]			COD [-]		
Sites	BC	$PM_{2.5}$	O_3	BC	$PM_{2.5}$	O_3
Canyon South - UTF	0.25	0.32	0.58	0.62	0.34	0.27
Canyon South - Roof	0.52	0.65		0.48	0.27	
Canyon South - Canyon North	0.57			0.31		
Canyon North - Roof	0.60			0.57		
Canyon North - UTF	0.23			0.68		
UTF - Roof	0.43	0.44		0.34	0.29	

Sunday morning (05:00–09:00) allowed the accumulation of O_3 to start earlier than on weekdays, resulting in higher O_3 levels.

3.3. Correlation analysis of key pollutants

In general, intrasite correlations between pairs of pollutants were higher than intersite correlations among individual pollutants. For example, BC at UTF showed weak spatio-temporal correlation with BC at the canyon sites (North and South) since the UTF site (located on the city's outskirts) receives very little urban influence (Tables 3 and 4). The BC correlation increased when considering the background sites (UTF vs. rooftop) and was highest between the urban sites. Krecl et al. (2016) showed results from a previous campaign conducted in Londrina in 2014 and concluded that the intersite BC correlation was highest during the early morning (00:00–04:00), when local sources weakened and BC levels became more homogeneous over the city. The moderate temporal correlation for BC between the North and South façades can be explained by the bus stop effect that largely increased the BC concentrations during the day at the North side (see Section 3.2). $PM_{2.5}$ concentrations showed a relatively low spatial variability when compared to BC, and temporal correlations were highest (0.65) between the canyon South and the rooftop site (Table 3). In general, O_3 levels did not vary much in space, but the temporal correlation was relatively low when considering all the hours of the day.

The highest R values ($R > 0.80$) were observed only at the canyon South site for BC, NOx and PN (Table 4), which are species mainly emitted by traffic exhausts in urban areas and displayed better correlation with the number of HDV on that street transect than with the total number of vehicles. Other emission sources than traffic exhaust might have important contribution to $PM_{2.5}$ over the city, thus causing less spatial variability (Table 3), and presenting a lower temporal correlation with BC, NOx and PN at the canyon South site (Table 4). Pérez et al. (2010) also found that the variability of $PM_{2.5}$ was less affected by direct traffic emissions when compared to BC, PN, NOx and other co-pollutants (CO and SO_2) in a study carried out in Barcelona, Spain. Kendrick et al. (2015) reported that $PM_{2.5}$ measurements conducted along an urban arterial in Portland, USA did not have a strong relationship with local traffic volumes, and concluded that they were more tied to regional sources and meteorological conditions.

3.4. Wind influence on pollutant concentrations

BC, PN, NOx, and NO_2 consistently decreased with increasing WS at all sites (Fig. 4), showing the positive effect of strong winds to disperse locally produced air pollution. Because in traffic-impacted environments, such as street canyons, a large fraction of PN consists of ultrafine particles (e.g., Krecl et al., 2015), changes in concentration are mainly found when the polluted air is diluted by fresh air transported at increasing WS (Kumar et al., 2008). The same underlying characteristics were reported by Jones et al. (2010) when analyzing the relationship between pollutant concentration (elemental carbon, PN, and NOx) and WS at three sites (canyon, urban background, and rural) in London. Krecl et al. (2015) also observed the same pattern for BC and NOx when studying the dependence of these pollutants on WS in Stockholm at three monitoring sites (street canyon, curbside, and rooftop).

In the case of $PM_{2.5}$, we observed that the mass concentration was diluted with increasing WS up to $5\,m\,s^{-1}$ (Fig. 4). Thereafter, resuspension processes and/or regional transport may have dominated and the $PM_{2.5}$ mass increased with WS. For example, road surface abrasion can generate particles in trafficked areas which can be resuspended by strong winds (Pant and Harrison, 2013). Because the slight increase in $PM_{2.5}$ levels was simultaneously observed at all sites when $WS > 5\,m\,s^{-1}$, it is most likely that this increase was connected to regional transport than to local processes in the city center.

Table 4
R between different pollutants and traffic rates measured at the canyon South site. $R > 0.80$ are displayed in bold face.

R [-]	BC	$PM_{2.5}$	NOx	NO_2	PN
$PM_{2.5}$	0.67				
NOx	**0.88**	0.56			
NO_2	0.74	0.60	**0.81**		
PN	**0.82**	0.59	**0.87**	0.67	
TR_{total}	0.36	0.29	0.36	0.29	0.46
TR_{HDV}	0.62	0.37	0.60	0.49	0.64

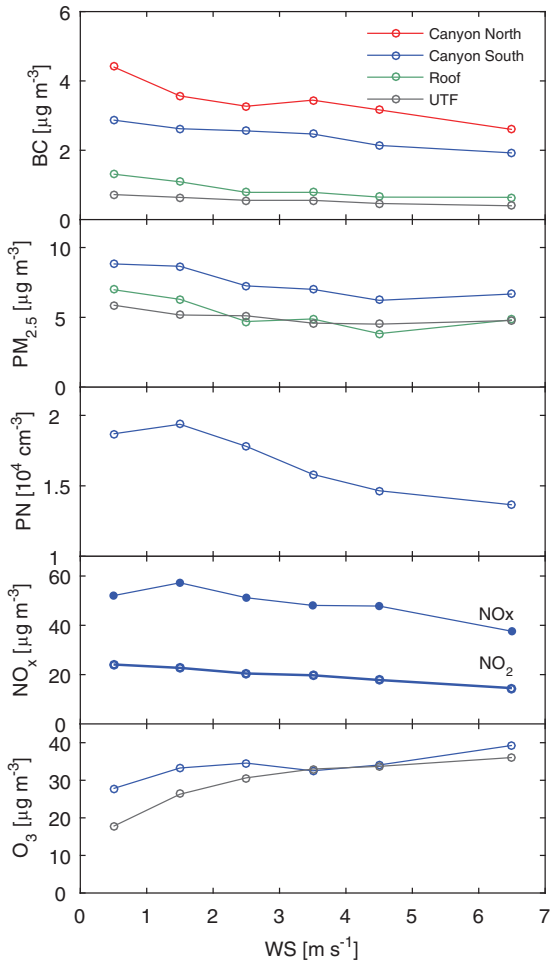

Fig. 4. Dependence of air pollutant concentrations on wind speed.

The O_3 concentrations also showed an upward trend with WS. We observed that the largest O_3 concentrations were associated with northerly and northeasterly winds and WS $> 5\,m\,s^{-1}$. This highlights the contribution of the regional transport of O_3 and/or its precursors, with the most likely upwind sources being polluted conurbations (such as the megacity of São Paulo) and biomass burning events, as shown by Krecl et al. (2016) in a previous study in the same region in 2014.

In relation to the WD dependence, we analyzed the mean concentration roses and presented them grouped per species (intersite comparison, Fig. 5a–c) and per site (intrasite comparison, Fig. 5e–g); the latter as normalized concentrations. Due to the presence of buildings on both sides of the canyon, rooftop winds blowing perpendicular to the street axis (± 30°) might trigger a recirculation vortex within the canyon that leads to higher pollutant concentrations at the leeward façade (Hunter et al., 1992). In this study, northerly (0 ± 30°) and southerly (180 ± 30°) winds represent cross-canyon flow directions for the leeward and windward situations (Fig. 5g). Concentration roses with asymmetrical patterns might be the result of local emission sources that affect pollutant levels in a different fashion depending on the WD, long-distance transport of pollutants, and/or variations of mean WS for the different wind sectors.

BC concentrations at the canyon sites were higher than at the background sites irrespective of the WD, and we confirmed a pronounced vortex effect on the concentrations measured at the South façade under southerly winds conditions and at the North side for northerly winds (Fig. 5a). Analyzing the concentration roses of BC street increments (street minus rooftop levels, not shown), the North façade presented a mean of $3.38\,\mu g\,m^{-3}$ for northerly winds, whereas the South façade experienced a mean value of $2.27\,\mu g\,m^{-3}$ for southerly winds. Since northerly and southerly winds occurred with comparable frequency and similar speeds (Fig. 5d), we can conclude that the mean difference of $1.11\,\mu g\,m^{-3}$ between the two façades was due to the influence of the bus stop.

In terms of absolute concentrations and regardless of the wind direction, the intersite difference for $PM_{2.5}$ is less evident than for BC (Fig. 5b). However, the vortex influence was still present in the canyon South data for southerly winds. The background sites presented similar patterns with higher $PM_{2.5}$ concentration for westerly winds, and maximum at the UTF site. This last feature could be associated with $PM_{2.5}$ advection from the city center and emissions in neighborhoods between the city center and the UTF campus (see Fig. 1), given that this pattern was not that accentuated at the rooftop. On average, O_3 concentrations at the canyon South and

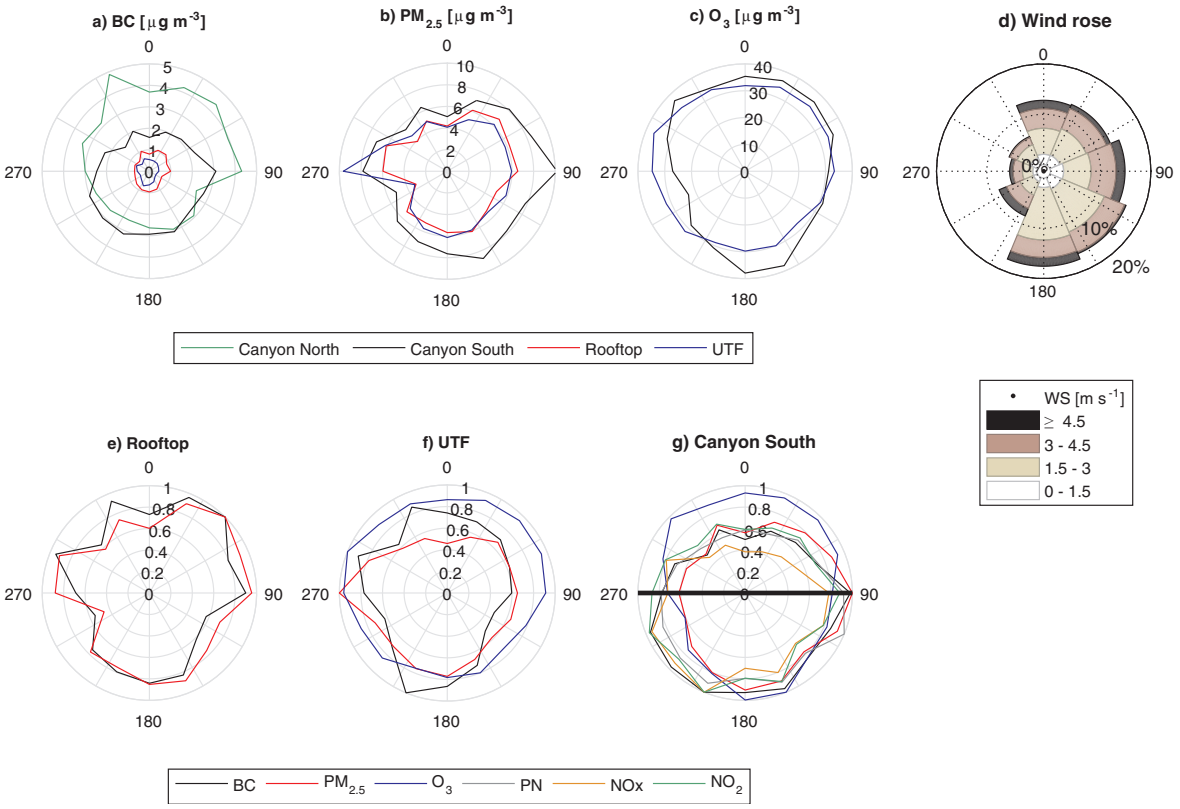

Fig. 5. Concentration roses of BC (a), $PM_{2.5}$ (b), and O_3 (c) together with wind rose (d) for the study period. Normalized concentration roses are shown for the rooftop (e), UTF (f) and canyon South (g) sites. The thick lines in panel g) represents the street canyon axis.

UTF showed little directionality, suggesting a more homogeneous distribution of the pollutant over the city.

From the intrasite analysis, the following features stand out: (i) $PM_{2.5}$ and BC showed the same pattern at the rooftop site whereas some differences were observed at the UTF campus, indicating the presence of local sources that contribute differently to BC and $PM_{2.5}$ in the suburban area, (ii) different patterns of concentration roses were observed at the canyon South site (Fig. 5g). O_3 showed a rather symmetrical distribution; BC, NOx, NO_2, and PN displayed higher concentrations for southerly winds due to the vortex recirculation, whereas $PM_{2.5}$ showed higher levels with easterly and southeasterly winds.

3.5. Urban vs. Regional contribution

3.5.1. PN and NOx urban background concentrations

The mean PN and NOx concentrations within the canyon on weekdays at 03:00 when the total TR was minimal (7 veh h^{-1}) were 2,803 cm^{-3} (road-to-roof ratio of 6.8) and 4.37 μg m^{-3} (ratio of 13.0), respectively. These results could largely underestimate the mean urban background concentrations, because traffic-related pollutants display a diurnal cycle at urban background sites connected to traffic activities within the city as depicted in Fig. 3 and in other works (Krecl et al, 2016; 2015; 2011).

A second option was to estimate the mean PN and NOx concentrations at the rooftop site by applying a ratio of 2.9 (mean BC ratio between South canyon and rooftop sites), which yielded 6,543 cm^{-3} and 19.38 μg m^{-3}, respectively. By doing this, we assumed that the strong correlation observed between the three pollutants at the canyon site (Table 4) might also apply to the rooftop location, and that these pollutants might disperse in the same fashion. However, other processes besides dilution might affect the particle dynamics during the transport towards the rooftop site. This was observed, for example, by Tuch et al. (2006) who reported a low correlation (lowest R = 0.3) between street canyon and urban background PN concentrations.

Several urban studies reported PN road-to-roof ratios between 1.9 and 3.0 (Tuch et al., 2006; Boogaard et al., 2010; Weber et al., 2013; Dos Santos-Juusela et al., 2013), whereas NOx ratios varied between 1.7 and 4.9 (Xie et al., 2003; Kwak et al., 2016 and references therein). This range of ratios can be attributed to several factors, such as traffic characteristics in the canyon, street canyon geometry, advection of pollutants from adjacent streets, wind and traffic-induced turbulences (Berkowicz, 2000). Thus, our approach to estimate the PN and NOx concentrations at the urban background site combines the use of the mean road-to-roof ratio found for BC in this study, and the minimum and maximum ratios reported in the literature for each species to evaluate their uncertainty.

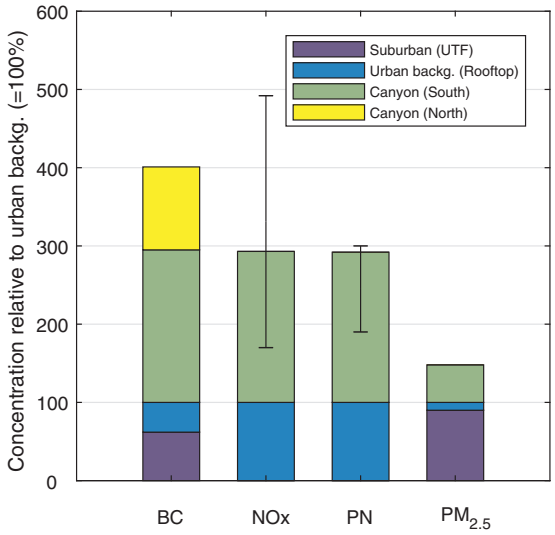

Fig. 6. Comparison of weekday concentrations at a suburban, urban background, and street canyon (both façades) sites relative to urban background concentrations. The error bars represent the uncertainty of NOx and PN contributions relative to the urban background.

3.5.2. Street and urban contributions to particulates and NOx

We now illustrate the contribution of the urban sources relative to the background levels and discuss the potential to decrease urban concentrations by reducing the local emissions of atmospheric pollutants. Fig. 6 displays the mean weekday concentration at the different sites relative to the concentrations at the urban background concentrations (rooftop equals to 100%). The concentration bars are stacked so that additional contributions are displayed. The error bars represent the uncertainty of NOx and PN contributions relative to the urban background.

For BC and $PM_{2.5}$, 38 and 10% of the background concentrations were generated in the urban area. Regarding the canyon South site, BC, NOx and PN concentrations were 195, (70–392), and (90–200) % relatively higher than urban background levels, whereas $PM_{2.5}$ was only 48% higher relative to the rooftop concentrations. Focusing on a microscale traffic environment, we estimated a reduction potential of 106% for BC concentrations at the North side compared to the South façade. If we consider that the BC advection from the north and south directions was roughly the same in the city center (Fig. 5a, rooftop), the possibility for reduction might be associated with urban planning actions, connected to the control of air pollution from motorized vehicles, relocation of bus stops and ideally even a new street configuration (to decrease the vortex recirculation).

Because diesel-powered buses presented a major influence on BC and PN concentrations in this study, the introduction of new emission standard stages with mandatory use of DPF and fleet renewal of buses with DPF should be priority actions to be conducted. To curb in-use emissions of existing vehicles, implementing I/M programs and retrofitting urban buses with DPF might be essential as it has been done in Santiago de Chile (SDC, 2011). These general strategies at city level could be reinforced by specific actions for the street canyon where we conducted our study, such as reducing the number of bus lines on that street to reach the bus terminal. This does not mean reducing the bus schedules, but splitting the access of the 22 bus lines into the city center over more streets to serve the bus terminal and, hence, cutting down the number of bus lines per street. Due to the large impact of the bus stop on the air quality when stop-and-go driving was predominant, we recommend that traffic engineers consider relocating bus stops placed within street canyons to more open areas to avoid exposing passengers, pedestrians and residents to elevated pollution levels. In our case, the bus stop could be relocated 150 m to the west on the same street where there is a square, with an unobstructed north façade. This might also help creating more evenly distributed bus stops on the canyon street where there are three stops currently (150 m between the first and second stops, and 650 m between the second and third stops).

3.5.3. Local/regional contribution to ozone

The scattergram of daylight mean OX vs. NOx is presented for the canyon South site (Fig. 7), where the offset and slope are the NOx-independent and the NOx-dependent contributions to O_3, respectively. The former represents the regional background O_3 level, while the latter is related to the O_3 production due to local sources. The NOx-independent contribution (24 ppbv, or 47.0 $\mu g\, m^{-3}$) was larger than the NOx-dependent contribution (40.6 $\mu g\, m^{-3}$ for the highest daylight mean value of NOx), which means that the potential for the local reduction of O_3 is lower than the regional contribution.

The regional contribution of O_3 associated with the transport of pollution plumes emitted by biomass burning is a well identified problem in several countries/regions around the world (e.g., Liu et al., 2013). In our study area, this problem might be even more severe during the dry season (August-October) when fires are more frequent for vegetation clearance and long-range transported pollutants affect the air quality of large and small cities downwind of the fire foci (e.g., Krecl et al., 2016; Lopes et al., 2012; Freitas et al., 2005). Besides anthropogenic biomass burning, other human activities developed in a large city might impact downwind cities through regional transport of O_3 as reported by Boián and Andrade (2012) and Krecl et al. (2016).

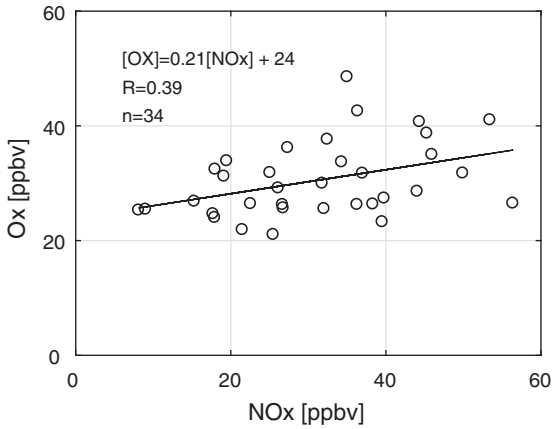

Fig. 7. Variation of daylight average mixing ratios of oxidant (OX) with the level of NOx. Data were calculated using daytime measurements only (between 08:00 and 17:00) The line was defined by regression analysis, and the correlation coefficient *R* and number of valid data *n* are also displayed.

4. Summary and conclusions

We investigated the concentrations of BC, O_3, NOx, $PM_{2.5}$ and PN in the urban core (street canyon and rooftop sites) and at a suburban site located on the outskirts of Londrina city in the period January 26th to March 27th 2016. As expected, the highest concentrations were registered in the city center at street level, except for O_3. Large hourly and daily variations were observed at the canyon sites, in contrast with the reduced variability found in the suburban area for the particulate pollutants. Concentrations of BC between sites were poorly correlated in time ($R < 0.70$), combined with highly heterogeneous concentrations (COD: 0.31–0.68) even at spatial scales as short as few meters within the canyon.

We identified that vehicle exhaust emissions, especially from the diesel fleet, largely influenced the BC, NOx and PN concentrations within the canyon. This conclusion is supported by the following key features: (i) the high correlation ($R > 0.80$) between hourly concentrations of BC, NOx and PN in the canyon suggests a common emission source; (ii) the moderate correlation between BC, NOx and PN and the TR of HDV, revealing the large impact of diesel bus emissions on ambient concentrations; (iii) the same pattern of BC, NOx and PN concentration roses was observed at the canyon South site; (iv) the three pollutants showed decreasing trends with increasing WS, suggesting that their emission sources were local.

The following facts indicate that $PM_{2.5}$ concentrations were impacted by a second emission source with regional origin: (i) the poor relationship of $PM_{2.5}$ concentrations with local traffic rates and with BC, NOx and PN concentrations at the canyon South site; (ii) the more homogenous distribution of $PM_{2.5}$ concentrations over the city (COD: 0.27–0.34); (iii) the increasing $PM_{2.5}$ concentrations with increasing WS at all sites, suggesting the contribution from regional transport; (iv) different shape of $PM_{2.5}$ concentration rose when compared to the other pollutants at the canyon South site, with the largest advection for easterly and south-easterly winds.

On average, O_3 levels were more homogeneous (COD: 0.27) over the city, but a closer inspection based on hourly concentrations revealed the “weekend effect” at the canyon site, particularly on Sundays when NOx concentrations were lower due to reduced TR. The raise in O_3 levels with increasing WS indicated regional transport, which was confirmed by analyzing the OX vs. NOx scattergram, yielding a regional contribution of 47.0 $\mu g\, m^{-3}$ and a maximum local contribution of 40.6 $\mu g\, m^{-3}$.

The potential for reducing pollutant concentrations was analyzed at different scales, and the results pointed to different abatement strategies. Concentrations of BC, PN, and NOx can be largely reduced in the city center by targeting the reduction of traffic emissions, particularly connected to the diesel fleet. At microscale level, reorganizing the traffic in street canyons either by changing the position of bus stops and/or reducing the number of buses circulating per street can be a relatively easy measure to improve the air quality in densely populated areas with intense commercial activity. Reducing $PM_{2.5}$ and O_3 concentrations is more challenging because of the substantial regional contribution observed for both species, and mitigation measures might include state, national or even transboundary agreements to abate these species and their precursors. We emphasize that air pollution regulations based solely on $PM_{2.5}$ mass concentrations might be insufficient to decrease the concentration of traffic-related particles such as BC and PN.

Finally, we highlight that the adopted methodology of measuring SLCP at key points in the city, even for a short period, can shed light on the possible strategies to reduce their emissions. The research outputs and the already proved suitable methodology can be applied to other medium-sized cities in Brazil where no air quality monitoring exists and there is an urgent need to assess the air pollution levels.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.trd.2019.01.032.

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